CLAIMS

1. A non-aqueous electrolyte for a lithium secondary battery to be used in combination with a positive electrode and a negative electrode capable of storing and releasing lithium, which comprises a non-aqueous solvent and a lithium salt dissolved therein,

wherein said non-aqueous solvent comprises:

(a) a phosphate comprising both (a1) a chain state phosphate and (a2) a cyclic phosphate; and

(b1) a cyclic carboxylate.

2. The non-aqueous electrolyte according to claim 1, wherein said chain state phosphate (a1) is contained in said non-aqueous solvent in an amount of 10 to 60% by volume, based on the total volume of said chain state phosphate (a1) and said cyclic carboxylate (b1).

3. A non-aqueous electrolyte for a lithium secondary battery to be used in combination with a positive electrode and a negative electrode capable of storing and releasing lithium, which comprises a non-aqueous solvent and a lithium salt dissolved therein,

wherein said non-aqueous solvent comprises:

(a) at least one phosphate selected from (a1) a chain state phosphate and (a2) a cyclic phosphate;

(b1) a cyclic carboxylate; and

at least one compound selected from (c1) a vinylene carbonate compound and (c2) a vinylethylene carbonate compound.

4. The non-aqueous electrolyte according to claim 3, wherein said phosphate (a) is contained in said non-aqueous solvent in an amount of 10 to 90% by volume, based on the total volume of said phosphate (a) and said cyclic carboxylate (b1).

5. A non-aqueous electrolyte for a lithium secondary battery to be used in combination with a positive electrode and a negative electrode capable of storing and releasing lithium, which comprises a non-aqueous solvent and a lithium salt dissolved therein,

wherein said non-aqueous solvent comprises:

(a) at least one phosphate selected from (a1) a chain state phosphate and (a2) a cyclic phosphate;

at least one compound selected from (c1) a vinylene carbonate compound and (c2) a vinylethylene carbonate compound; and

at least one compound selected from the group consisting of (d1) a cyclic amide compound, (d2) a cyclic carbamate compound, and (d3) a heterocyclic compound.

6. The non-aqueous electrolyte according to claim 5, wherein said non-aqueous solvent further comprises (b1) a cyclic carboxylate, wherein said phosphate (a) is contained in said non-aqueous solvent in an amount of 10 to less than 100% by volume, based on the total volume of said phosphate (a) and said cyclic carboxylate (b1).

7. A non-aqueous electrolyte for a lithium secondary battery to be used in combination with a positive electrode and a negative electrode capable of storing and releasing lithium, said non-aqueous electrolyte comprising a non-aqueous solvent and a lithium salt dissolved therein

wherein said non-aqueous solvent comprises:

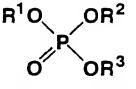
(a) at least one phosphate selected from (a1) a chain state phosphate and (a2) a cyclic phosphate; and

(c1) a vinylene carbonate compound and (c2) a vinylethylene carbonate compound.

8. The non-aqueous electrolyte according to claim 7, wherein said non-aqueous solvent further comprises at least

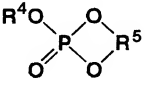
one compound selected from (b1) a cyclic carboxylate and (b2) a cyclic carbonate, wherein said phosphate (a) is contained in said non-aqueous solvent in an amount of 60 to less than 100% by volume, based on the total volume of said phosphate (a) and said at least one compound selected from the cyclic carboxylate (b1) and the cyclic carbonate (b2).

9. The non-aqueous electrolyte according to any one of claims 1 to 8, wherein said chain state phosphate (a1) is represented by said following formula (I):



(I)

wherein R^1 to R^3 each independently represent an unsubstituted or fluorine-substituted linear or branched alkyl group having 1 to 4 carbon atoms, and said cyclic phosphate (a2) is represented by the following formula (II):



(II)

wherein R^4 represents an unsubstituted or fluorine-substituted, linear or branched alkyl group having 1 to 4 carbon atoms, and R^5 represents a linear or branched alkylene group having 2 to 8 carbon atoms.

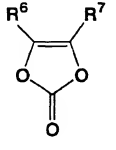
10. The non-aqueous electrolyte according to claim 9, wherein said chain state phosphate (a1) is at least one chain state phosphate selected from the group consisting of trimethyl phosphate, trifluoroethyldimethyl phosphate, bis(trifluoroethyl)methyl phosphate and tris(trifluoroethyl) phosphate, and said cyclic phosphate (a2) is at least one cyclic phosphate selected from the

group consisting of ethylenemethyl phosphate, ethyleneethyl phosphate and ethylenetrifluoroethyl phosphate.

11. The non-aqueous electrolyte according to any one of claims 1, 3, 6 and 8, wherein said cyclic carboxylate (b1) is at least one cyclic carboxylate selected from the group consisting of γ-butyrolactone, γ-valerolactone, γ-caprolactone, γ-octanolactone, β-butyrolactone, δ-valerolactone, and ε-caprolactone.

12. The non-aqueous electrolyte according to claim 8, wherein said cyclic carbonate (b2) is at least one cyclic carbonate selected from the group consisting of ethyelne carbonate, propylene carbonate, and butylene carbonate.

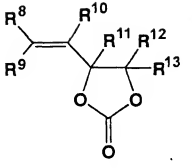
13. The non-aqueous electrolyte according to any one of claims 3, 5 and 7, wherein said vinylene carbonate compound (c1) is represented by the following formula (III):



(III)

wherein R^6 and R^7 each independently represent a hydrogen atom, an alkyl group having 1 to 4 carbon atoms, or a branched alkyl group,

and said vinylethylene carbonate compound (c2) is represented by the following formula (IV):



(IV)

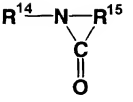
wherein R^8 to R^{13} each independently represent a hydrogen atom, or a linear or branched alkyl group having 1 to 4 carbon atoms.

14. The non-aqueous electrolyte according to claim 13, wherein said vinylene carbonate compound (c1) is at least one vinylene carbonate compound selected from the group consisting of vinylene carbonate, 4-methylvinylene carbonate, 4-ethylvinylene carbonate, 4,5-dimethylvinylene carbonate, 4,5-diethylvinylene carbonate and 4-methyl-5-ethylvinylene carbonate, and said vinylethylene carbonate compound (c2) is at least one vinylethylene carbonate compound selected from the group consisting of 4-vinylethylene carbonate, 4-vinyl-4-methylethylene carbonate, 4-vinyl-4-ethylethylene carbonate, 4-vinyl-4-n-propylethylene carbonate, 4-vinyl-5-methylethylene carbonate, 4-vinyl-5-ethylethylene carbonate, and 4-vinyl-5-n-propylethylene carbonate.

15. The non-aqueous electrolyte according to claim 13, wherein a content of at least one compound selected from said vinylene carbonate compound (c1) and said vinylethylene carbonate compound (c2) is 0.1 to 15% by weight based on the total weight of said non-aqueous electrolyte.

16. The non-aqueous electrolyte according to claim 5 or 6, wherein said cyclic amide compound (d1) is represented by

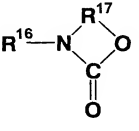
the following formula (V):



(V)

wherein R^{14} represents a linear or branched alkyl group having 1 to 4 carbon atoms, a vinyl group or an allyl group, or a cycloalkyl group, an aryl group or an aralkyl group having 6 to 8 carbon atoms, and R^{15} represents a divalent hydrocarbon group having 2 to 8 carbon atoms,

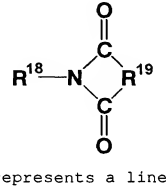
said cyclic carbamate compound (d2) is represented by the following formula (VI):



(VI)

wherein R^{16} represents a linear or branched alkyl group having 1 to 4 carbon atoms, a vinyl group or an allyl group, or a cycloalkyl group, an aryl group or an aralkyl group having 6 to 8 carbon atoms, and R^{17} represents a divalent hydrocarbon group having 2 to 8 carbon atoms,

and said heterocyclic compound (d3) is represented by the following formula (VII):



(VII)

wherein R^{18} represents a linear or branched alkyl

group having 1 to 4 carbon atoms, a vinyl group or an allyl group, or a cycloalkyl group, an aryl group or an aralkyl group having 6 to 8 carbon atoms, and R^{19} represents a divalent hydrocarbon group having 2 to 8 carbon atoms.

17. The non-aqueous electrolyte according to claim 16, wherein a content of said at least one compound selected from the cyclic amide compound (d1), the cyclic carbamate compound (d2) and the heterocyclic compound (d3) is 0.1 to 15% by weight based on the total weight of the non-aqueous electrolyte.

18. The non-aqueous electrolyte according to any one of claims 1, 3, 5 and 7, wherein said lithium salt is an inorganic acid lithium salt selected from $LiPF_6$ and $LiBF_4$, or an organic acid lithium salt selected from the group consisting of $LiCF_3SO_3$, $LiN(CF_3SO_2)_2$, $LiN(C_2F_5SO_2)_2$, $LiN(CF_3SO_2)(C_4F_9SO_2)$, $LiPF_3(C_2F_5)_3$ and $LiB(CF_3COO)_4$.

19. A lithium secondary battery comprising the non-aqueous electrolyte according to any one of claims 1, 3, 5 and 7, and a positive electrode and a negative electrode which are capable of storing and releasing lithium.

20. The lithium secondary battery according to claim 19, wherein said negative electrode satisfies the following conditions:

(1) said negative electrode comprises an anode material comprising a graphite carbonaceous material (A) having a plane spacing d_{002} value of the (002) plane of less than 0.337 nm and a carbonaceous material (B) having the plane spacing d_{002} value of the (002) plane of 0.337 nm or more, as measured by wide-angle X-ray diffractometry;

(2) that the weight ratio between said graphite carbonaceous material (A) and said carbonaceous material

(B) is 99.5:0.5 to 50:50; and

(3) that said anode material has an R value of more than 0.2 and 1.5 or less, wherein the R value is represented by IB/IA wherein IA represents a peak intensity appearing in the range of from 1,570 to 1,620 cm^{-1}, and IB represents a peak intensity appearing in the range of from 1,350 to 1,370 cm^{-1}, as measured by Raman spectroscopy using an argon ion laser with a wavelength of 514.5 nm.

21. The lithium secondary battery according to claim 20, wherein said graphite carbonaceous material (A) has at least part of a surface thereof coated with said carbonaceous material (B).

22. The lithium secondary battery according to claim 20, wherein said anode material is obtained by calcining a mixture of said graphite carbonaceous material (A) and an organic material.

23. The lithium secondary battery according to claim 22, wherein said calcination is conducted at a calcination temperature of 500 to 2,200°C.

24. The lithium secondary battery according to claim 20, wherein the R value of said anode material is 0.35 to 1.1.

25. The lithium secondary battery according to claim 20, wherein the R value of said anode material is 0.4 to 0.9.

26. The lithium secondary battery according to claim 20, wherein said anode material has an intensity ratio represented by ABC(101)/AB(101) of 0.15 or more, wherein AB(101) represents a peak intensity ascribed to the orientation of the hexagonal crystal system graphite layer, and ABC(101) represents a peak intensity ascribed to the orientation of the rhombohedral crystal system graphite

layer, as measured by wide-angle X-ray diffractometry.

27. The lithium secondary battery according to claim 20, wherein said graphite carbonaceous material (A) has an intensity ratio represented by ABC(101)/AB(101) of 0.2 or more.

28. The lithium secondary battery according to claim 20, wherein said anode material comprising said graphite carbonaceous material (A) and said carbonaceous material (B) has a surface area of 0.5 to 25 m^2/g as measured by a BET method.

29. The lithium secondary battery according to claim 20, wherein said anode material comprising said graphite carbonaceous material (A) and said carbonaceous material (B) has a particle diameter of 4 to 40 μm.

30. The lithium secondary battery according to claim 19, wherein said negative electrode comprises at least one anode material selected from a carbonaceous material having a d value of the (002) plane of 0.335 to 0.34 nm as measured by X-ray diffractometry, an oxide of at least one metal selected from Sn, Si, and Al, and an alloy of lithium and at least one metal selected from Sn, Si, and Al.